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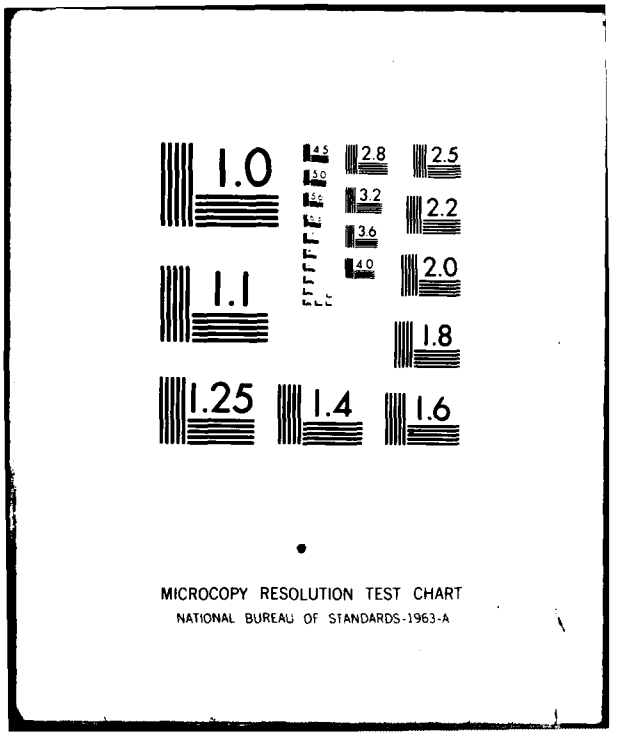
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THERMOELECTRIC MATERIALS FOR SOLAR ENERGY CONVERSION

BY J. F. GOFF J. R. LOWNEY

RESEARCH AND TECHNOLOGY DEPARTMENT

1 FEBRUARY 1979

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The thermoelectric efficiency index of N-type β-SiC has been calculated by use of the Goff-Lowney integral formulation. The scattering parameters were estimated by fitting the thermoelectric power and electrical conductivity data of Golikova, et al. The parasitic photon thermal conductivity was calculated theoretically by use of the theory of Devyatkova, et al, while the lattice thermal conductivity was treated parametrically. The results indicate that there exists an optimum carrier concentration of approximately 2 x 10^{20} cm^{-3} and that the efficiency is still increasing at 2000K.

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SUMMARY

The thermoelectric efficiency index of N-type β-SiC has been calculated by use of the Goff-Lowney integral formulation. The scattering parameters were estimated by fitting the thermoelectric power and electrical conductivity data of Golikova et al. The parasitic photon thermal conductivity was calculated theoretically by use of the theory of Devyatkova et al while the lattice thermal conductivity was treated parametrically. The results indicate that there exists an optimum carrier concentration of approximately 2 x 10^{20} cm^{-3} and that the efficiency is still increasing at 2000K.

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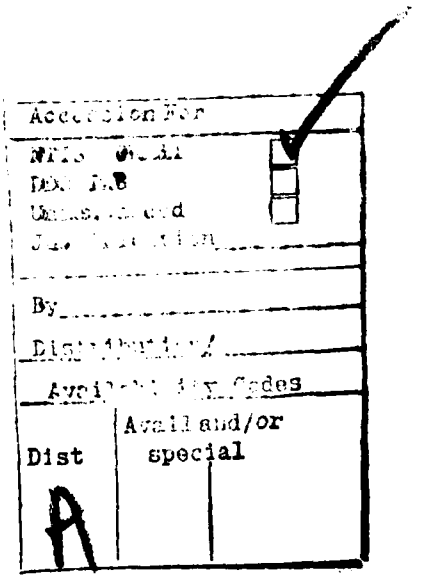

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ILLUSTRATIONS

I. INTRODUCTION

We have been asked to consider the potential of SiC as a material for a high temperature, solar thermoelectric generator. To do so we have analyzed the thermoelectric figure of merit, Z, with the aid of a special computer program. This quantity, or rather ZT, where T is the absolute operating temperature, is used as a measure of the efficiency of a thermoelectric material. Generally one wishes ZT to be greater than unity.

The quantity ZT is given by:

$$ZT = \frac{S^2 \sigma T}{K} \tag{1}$$

Where S is the thermoelectric power, σ is the electrical conductivity, and K is the total thermal conductivity. These quantities are not independent of one another, and Eq. 1 may be reformulated[1] to yield the more fundamental form, valid for cubic symmetry:

$$ZT = (p(e^2 T K_p/M_2+1) - 1)^{-1} \tag{2}$$

where

$$p = \frac{M_o M_2}{M_1^2} \tag{3}$$

and

$$M_n = - \int \sigma (E) E^n \frac{\partial f}{\partial E} dE \tag{4}$$

The M_n integrals are energy moment integrals of the conductivity over the Fermi distribution. K_p is the thermal conductivity due to all nonelectronic, parasitic mechanisms such as phonons and photons. In this formalism the ambipolar thermal conductivity is included in the M_2 integral.

In general for efficient material, it is necessary to minimize both p and the ratio TK_p/M_2. These two conditions lead usually to a trade-off and therefore to a region of optimum performance for any material depending on its properties. Minimization of p requires reducing the Fermi energy as much as possible, by

[1]Goff, J. F. and Lowney, J. R., Trans. Ar. Nuc. Soc. 23, 121 (1976).

lowering the carrier density or raising the temperature, so long as minority carrier effects remain negligible. However, the minimization of the TK_pM_2 ratio requires low photon and lattice phonon thermal conductivities with respect to M_2/e^2T. Achieving this condition further restricts the operating temperature and carrier density of the material. It may also require that the lattice phonon thermal conductivity be lowered by alloying or doping the pure material.

Evaluation of ZT for a particular material requires a knowledge of the band structure, electronic scattering processes, and nonelectronic thermal conductivity of the material. A comprehensive review of the SiC literature was supplied to us by the Solar Energy Research Institute. However, despite the completeness of the search, the quality and quantity of the available data are not adequate to answer all the questions concerning the thermoelectric behavior of the material. The primary reason for this inadequacy is that all the data has not been measured on the same samples. Therefore estimates will have to be made based broadly on what data is available.

SiC is a wide band-gap semiconductor which exists in three crystal types (cubic, hexagonal, and rhombohedral) as well as in many polymorphisms for the two non-cubic types. Each polymorph has its own set of properties, which may be very anisotropic for high order polymorphs.[2] The most common polytype is 6H, a relatively low order hexagonal structure. Most of the data on SiC correspond to this polytype. However, even for 6H, a definitive data set does not exist since the electrical and thermal properties were not measured on the same sample. The data indicate that both electrical and thermal conductivities vary substantially from sample to sample.[3] At best, estimates can be made on the basis of a collection of data from various sources.

The band structure of SiC is only fairly well known. The cubic and 2H forms have been calculated, but the higher polymorphs of hexagonal and rhombohedral forms have not been done because of their complexity. However, optical data show that the energy gap is large for all polytypes. In fact it is at least 10 kT even up to 2000K. Therefore intrinsic carrier densities should be small even at high temperature, although they may start to be appreciable above 2000K.

Carriers are introduced into SiC by adding impurities. A common donor is nitrogen; a common acceptor is aluminum. The impurity level energies and densities of states are not well enough known. We shall assume shallow impurity states with the activation energies given in several papers.[3] We expect that at high impurity densities, these states coalesce with the band and form a set of conducting states.[4] Thus the carrier density equals the impurity density as if all the states were ionized.

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[2]Lomakina,G.A. and Vodakov, Yu. A., Sov. Phys. Solid State 15, 83 (1973).

[3]Neuberger, M. "Electronic Properties Information Center Report on SiC," Hughs Aircraft, Culver City, CA (1965).

[4]Chao, K. A., Oliverira, F. A., and Majlis, N., Solid State Commun. 21, 845 (1977).

The scattering of carriers is also not fully understood. However, it may be assumed that the dominant mechanisms are ionized impurity scattering and lattice phonon scattering. The SiC data exhibit similar behavior to that of Ge, for which these two mechanisms dominate.

The thermal conductivity data set is incomplete as well. It does not extend over a wide enough range of carrier densities to determine the reduction of lattice thermal conductivity due to doping. Furthermore, none of the data give the orientation of the crystals associated with the measurements. The anisotropic crystals should have anisotropic thermal conductivities, and these anisotropies must be known for the calculations of non-cubic crystals.

We shall focus on the case of cubic (β) SiC for which we have already developed the necessary formalism. To compute properties of the other crystal structures, a full tensor formulation would be needed. A sufficient data set exists to estimate the ZT product of cubic SiC for various lattice thermal conductivities. Our procedure was to fit the resistivity and thermoelectric power data of Golikova, et al.[5] for n-type cubic SiC heavily doped with nitrogen. We have been able to fit their data much better than they were since we included both ionized impurity and lattice phonon scattering throughout the temperature range (300-2000K).

The model obtained by this procedure allowed us to calculate ZT as a function of carrier density, lattice thermal conductivity, and temperature. The thermal conductivity due to photons was also calculated and shown to be negligible at these high carrier densities for temperatures even as high as 2000K. As a result, SiC can operate at much higher temperature than is usually the case. The results of our calculations show also that for efficient operation, the lattice thermal conductivity must be reduced from about 100 to at least 10 mw/cmK. This may be accomplished by alloying or doping. Furthermore, efficient operation constrains the carrier density to be in the low 10^{20}cm^{-3} range, and the temperature to be close to 2000K or somewhat higher. Finally, we discussed the potential of several other materials for solar thermal energy conversion.

II. DATA ANALYSIS

We have analyzed the data of Golikova et al.[5] for n-type β-SiC heavily doped with nitrogen. They measured the electrical conductivity and thermoelectric power between 300 and 2000K in the carrier density range 3.5 to 8.0 x 10^{20} cm^{-3}. We expect that their lowest reported carrier density of 2.2 x 10^{20} cm^{-3} is in error and should be 3.5 x 10^{20} cm^{-3} to be consistent with the other two measured carrier densities and their corresponding mobilities. Since Hall voltages are small at these high carrier densities, small voltage errors can lead to substantial differences in inferred carrier density.

These data for S and σ are shown in Figures 1 and 2, respectively. The data for S have been replotted as log-log to more clearly delineate their temperature dependence at high temperatures.

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[5]Golikova, O.A., Ivanova, L.M., Pletyoshkin, A.A., and Semenenko, V.P., Sov. Phys. Semiconductors 5, 366 (1971).

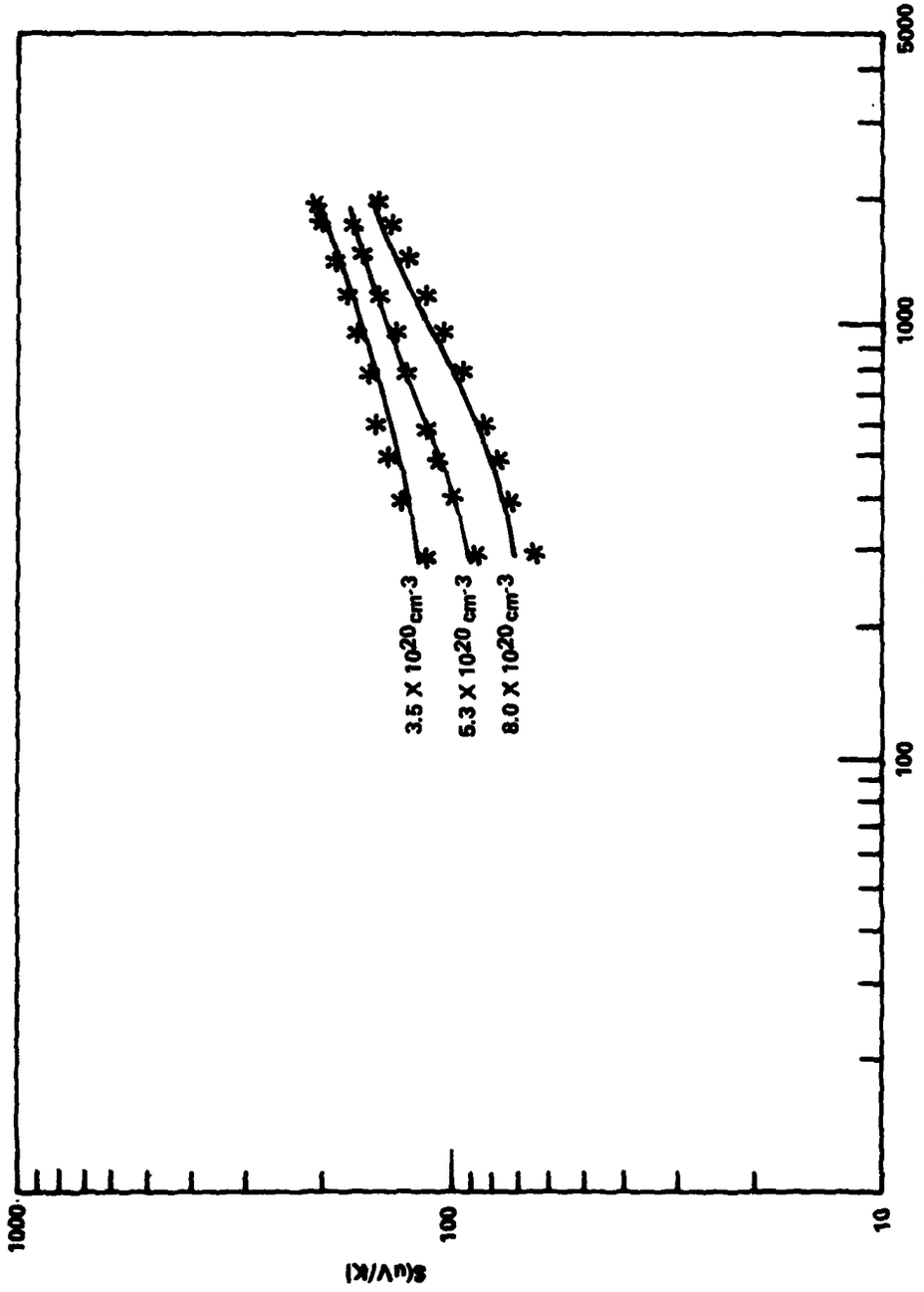

FIGURE 1 THE THERMOELECTRIC POWER X vs. TEMPERATURE T FOR N-TYPE, CUBIC SiC. THE POINTS HAVE BEEN COMPUTED FROM BASIC SCATTERING MECHANISMS

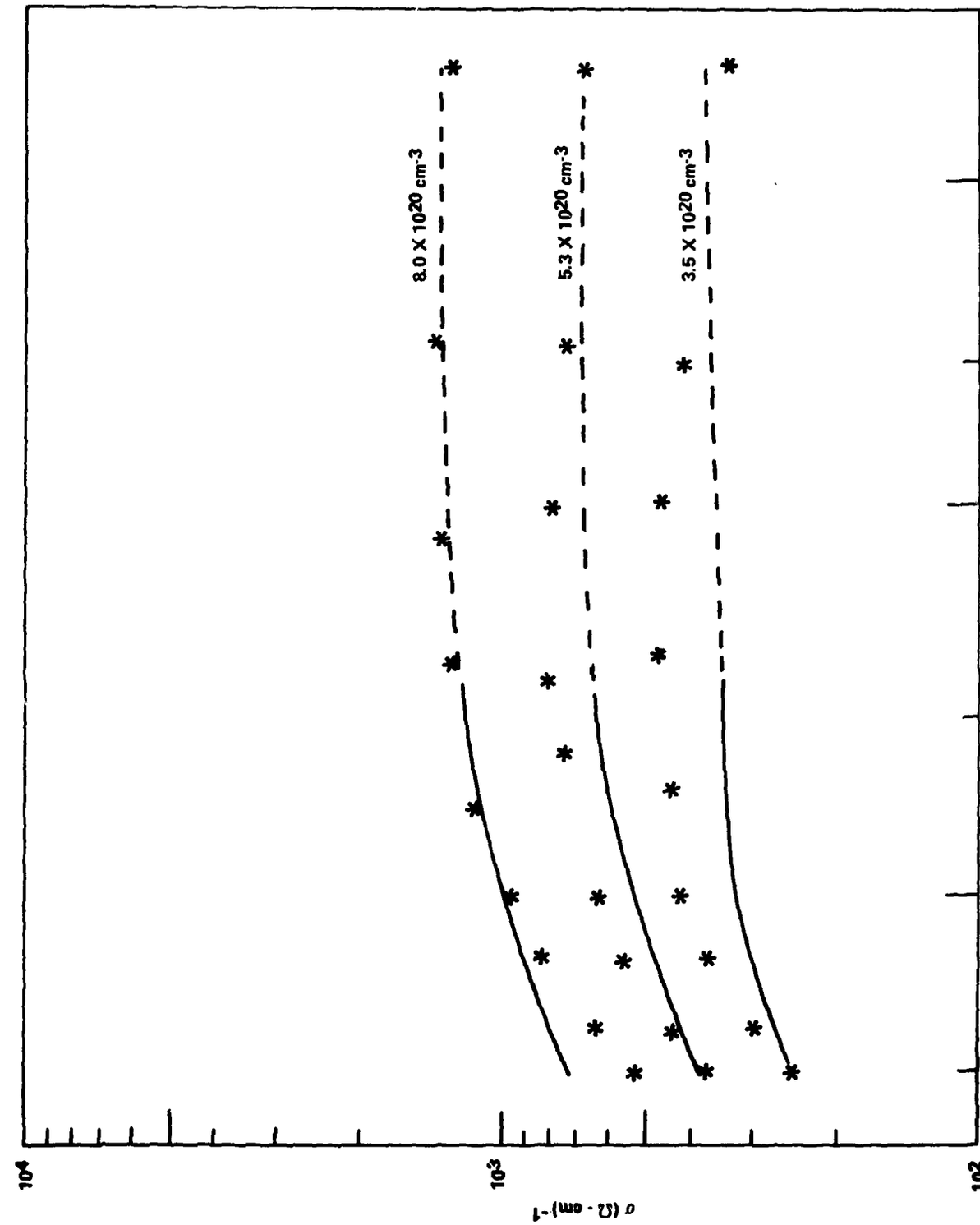

FIGURE 2 THE ELECTRICAL CONDUCTIVITY vs. $10^3/T$ FOR N-TYPE, CUBIC SiC. THE POINTS HAVE BEEN COMPUTED FROM BASIC SCATTERING MECHANISMS

The fitting procedure was accomplished in stages. Golikova et al.[5] were unable to fit their data because they assumed that there was only one scattering mechanism, ionized impurity scattering, which was dominant throughout their temperature range. They discuss the need to include lattice scattering at the highest temperatures but invoke an unrealistically high temperature dependence for it to do so. We have been able to accomplish a fit to both the resistivity and thermoelectric power by combining the effects of ionized impurity and lattice phonon scattering throughout the temperature range. The development of this model was suggested by the similar behavior of degenerate Ge.[6] Ionized impurity scattering dominates at low temperature, whereas lattice phonon scattering dominates at high temperature as expected. At intermediate temperatures both mechanisms contribute.

It should be remarked that the fitting of this data is complicated by the awkward temperature range and its own incompleteness. Since the Debye temperature θ of SiC is 1200 K, [7] one might expect that the relaxation time approximation would breakdown at approximately $\theta/2$ or 600 K. Such a conclusion is not warranted because small carrier concentrations such as found in these samples do not interact with the complete Debye sphere but only its inner portions[8] which randomize at lower temperatures. Nevertheless, both the slope and the magnitude of the ideal electrical resistivity ρ_i become functions of the carrier concentration n as well as the temperature. Further, a careful analysis of Golikova's σ data indicate that they did not measure to a low enough T to define the residual resistivity ρ_0. Thus, the exact nature of the impurity scattering, the electron-phonon interaction, and the balance between them is uncertain.

We assumed a parabolic band with an effective mass which decreases slightly with temperature. This decrease is expected since the energy gap also decreases slightly with temperature.[3] For best results, we found that the band mass should be given by:

$$m^*=0.58\ (1-1.0\times10^{-4}\ (T-300)) \tag{5}$$

which is a variation of less than half as much as that of the energy gap. At room temperature the effective mass m^* is 0.58, very close to the value of 0.60 reported in the literature for low carrier density samples.[3] This agreement implies that the energy band is not distorted by the coalescence with the impurity states. It appears that the donor electrons simply fill the band states and form a very degenerate semiconductor.

The electron scattering is assumed to be characterized by a relaxation time $\tau(E)$. For ionized impurity scattering $\tau(E)$ is given by:

$$\tau(E) \propto E^{3/2} \tag{6}$$

For lattice phonon scattering $\tau(E)$ is given by:

$$\tau(E) \propto E^{-1/2} \tag{7}$$

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[6]Goff, J. F. and Pearlman, N., Phys. Rev. 140, A2151 (1965).

[7]Prevot, B. and Carabatos, C., C. R. Acad. Sc. Paris 1 273 (1971).

[8]Wilson, A. H., "The Theory of Metals" Cambridge University Press, 1954, pp. 254, 278.

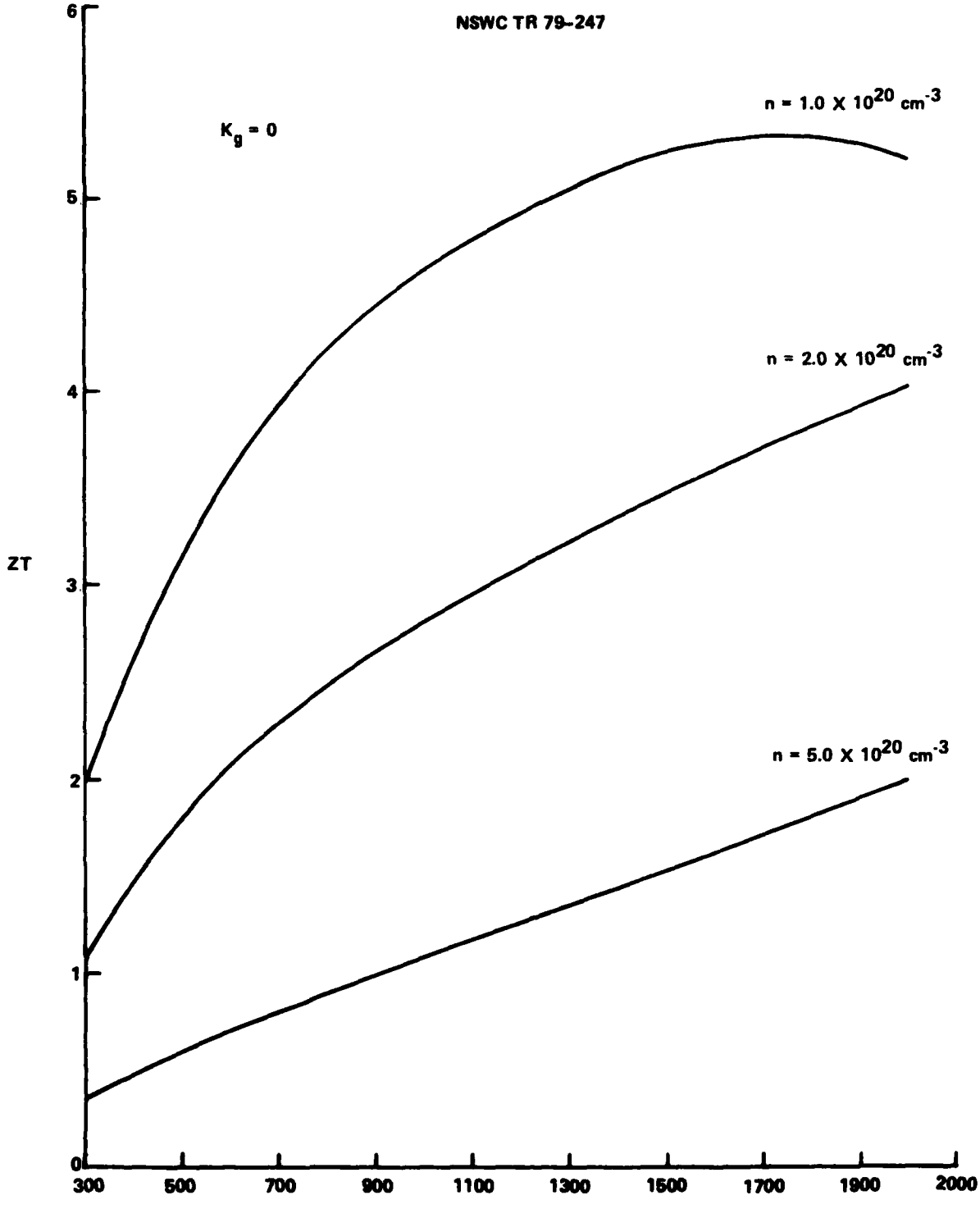

FIGURE 3 THE THERMOELECTRIC EFFICIENCY ZT vs. TEMPERATURE T AND CARRIER DENSITY n FOR LATTICE THERMAL CONDUCTIVITY $K_g = 0$. THE THEORETICAL PHOTON THERMAL CONDUCTIVITY K_r HAS BEEN INCLUDED IN THE CALCULATIONS

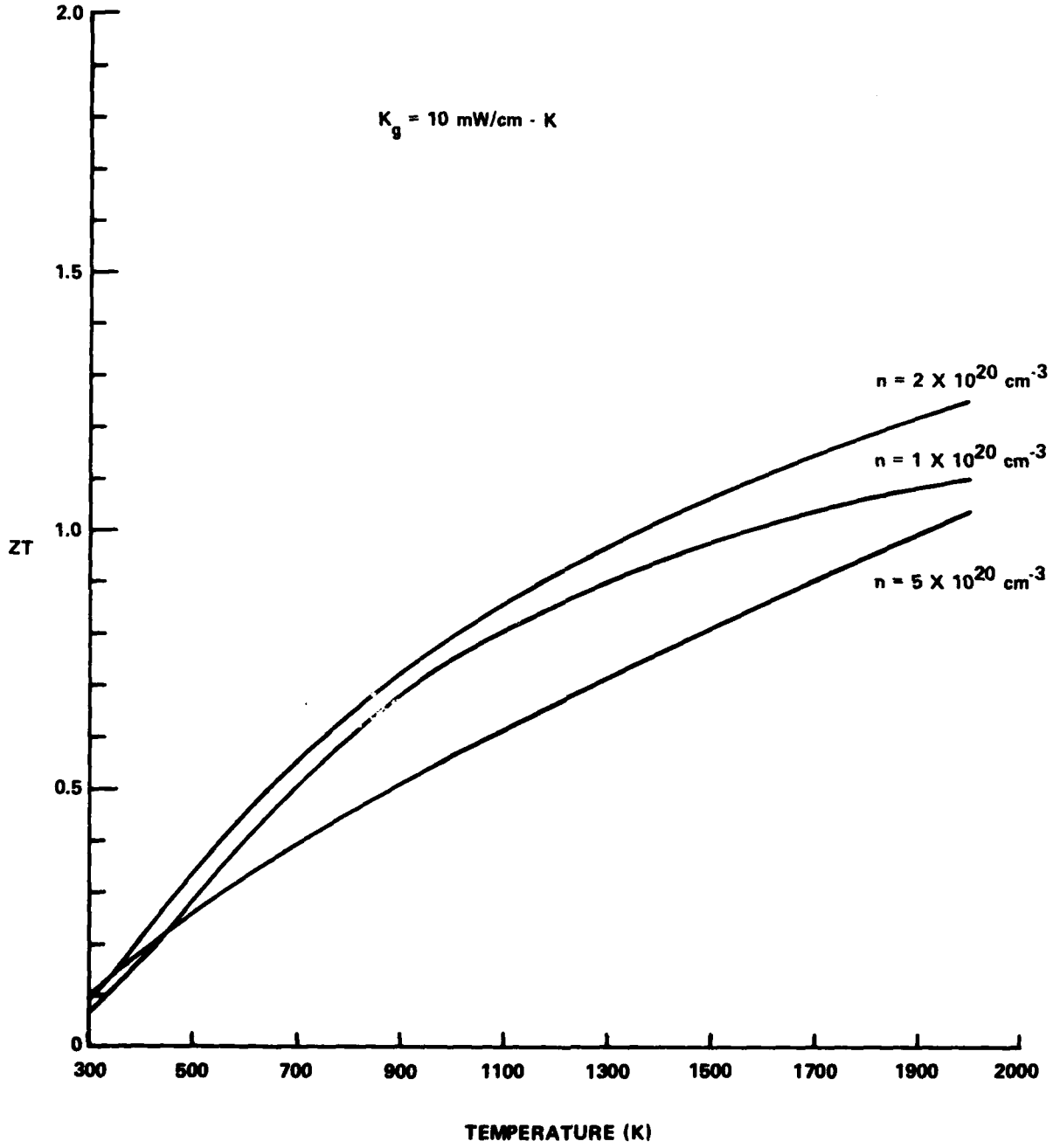

FIGURE 4 THE THERMOELECTRIC EFFICIENCY ZT vs. TEMPERATURE T AND CARRIER DENSITY n FOR A LATTICE THERMAL CONDUCTIVITY K_g = 10.0 mW/cm-K

The thermoelectric power S depends only on the energy dependence of the scattering, and not on its absolute magnitude. It was found that near 300K, ionized impurity scattering alone yielded a good fit to the data. Near 2000K, phonon scattering alone was satisfactory. To fit the data in between both mechanisms must be included. The temperature dependencies and magnitudes of these mechanisms were established by fitting the thermopower and conductivity data simultaneously. The results are:

$$\text{Ionized Impurity:} \quad \tau(E) = 1.16 \times 10^{-16}\, TE^{3/2} \text{ sec/K(eV)}^{3/2} . \tag{8}$$

$$\text{Lattice Phonon:} \quad \tau(E) = 1.53 \times 10^{-12}\, T^{-1}\, E^{-1/2} \text{ sec K(eV)}^{1/2} \tag{9}$$

The calculations of S and σ which were made with these relaxation times are shown by the asterisks in Figures 1 and 2, respectively. The fit to S is rather good over the whole temperature range. The fit to σ is adequate in the temperature range of interest; that is, at the higher temperatures. Having modeled the electronic behavior of the material, we were able to calculate the M_n integrals needed for ZT.

III. ZT PROJECTIONS

The evaluation of ZT requires values of both the M_n integrals already calculated and the parasitic thermal conductivity. The parasitic thermal conductivity, K_p is a sum of two terms: (1) lattice phonon thermal conductivity, K_g, and (2) radiative photon thermal conductivity, K_r. The first term depends on alloying, doping, and other material impurities and defects. We are treating K_g as an adjustable parameter in order to determine what its value must be for efficient operation. The second term may be calculated according to the analysis of Devyatkova et al.[9] which is outlined in Appendix A.

The carrier densities are so high in the samples we have fit ($>10^{20}\text{cm}^{-3}$) that the photon thermal conductivity is less than 1.0 mW/cmK even at temperatures as high as 2000K. For most materials, 1500K is an upper limit of efficient operation because of photon thermal conduction. We have included the value of K_r in K_p in our calculation of ZT, but it is K_g rather than K_r which limits performance. The very small valve of K_r allows SiC to be used at temperatures well above the usual limit.

The plots of ZT as a function of carrier density, n, lattice thermal conductivity, K_g, and absolute temperature, T, are shown in Figures 3 and 4. In Figure 3, K_g = 0.0; and ZT values in excess of 5.0 are possible at a carrier density of 1.0×10^{20} cm^{-3}. The small reduction in ZT for this carrier density above 1800K is caused by K_r, which is increasing with T and starting to become important. For the higher carrier densities, 2.0×10^{20} cm^{-3} and 5.0×10^{20} cm^{-3}, ZT decreases since the Fermi energy resides higher in the band.

In Figure 4, K_g = 10.0mW/cm-K; and the behavior of ZT is very different. The effect of K_r us negligible compared with K_g. The values of ZT are all lower,

[9]Devyatkova, E.D., Moizhes, B.Ya., and Smirnov, I.A., Sov. Phys. Solid State 1, 55 (1959).

reaching 1.25 for $n = 2 \times 10^{20}$ cm^{-3} at 2000K. For both lower and higher carrier densities, ZT is lower. At 1.0×10^{20} cm^{-3}, ZT is lower because of a smaller value of M_2. At 5.0×10^{20} cm^{-3} ZT is lower because the value of p is smaller. Therefore there exists an optimum carrier density. Furthermore, operation is best at high temperatures.

The question arises as to whether Golikova's data indicate minority carrier effects at temperatures on the order of 2000K. One would expect minority carrier effects to be more apparent in the behavior of S than in σ for two reasons: S depends upon a higher order moment integral and so is more sensitive to conduction contributions far from the Fermi energy than is σ; secondly, since S depends upon the sign of the carriers, minority carriers being opposite in sign to the majority ones and tend to cancel the thermoelectric power.

Golikova plotted S on a Cartesian plot, and, indeed S appears to be approaching a maximum. However the log-log plot of Figure 1 indicates that this maximum is only apparent and is in fact an indication that S has a $T^{0.4}$ dependence. Thus, these data do not unequivocally indicate minority carrier effects even at 2000K.

It is important to note the necessity for relatively high carrier densities in optimal SiC. The carriers are needed to produce a large M_2 value since the mobility of the carriers is low in SiC. The reason that the mobility is low is that the effective mass is large, as expected for this wide band-gap semiconductor. In narrow gap material, the mobility is characteristically higher; and optimum performance is achieved for much lower carrier densities and temperatures.

IV. CONCLUSION

We have fit the electrical conductivity and thermoelectric power data of Golikova at al.[5] and have obtained a model of electronic conduction for their high carrier density n-type cubic SiC. The thermoelectric efficiency index ZT was calculated as a function of carrier density and temperature by using this model and treating K_g, the lattice thermal conductivity, as an adjustable parameter. The photon thermal conductivity K_r was calculated as well and shown to be less than 1.0mW/cmK for carrier densities above 1.0×10^{20} cm^{-3} and temperatures below 2000K. Optimally efficient operation ($ZT \geq 1$) was shown to occur at high temperatures (2000K) for carrier densities in the low 10^{20} cm^{-3} range and K_g less than 10mW/cmK.

Achieving efficient performance will require lowering K_g an order of magnitude from its value of 100 mW/cmK for pure material at 2000K.[10] This may be possible by alloying or doping. The effects of such an effort on the electrical characteristics will have to be considered as well. However, the fact that the carrier density is so high will probably allow for considerable alteration of K_g without lowering the mobility appreciably.

Thus SiC is a wide band-gap semiconductor which may be applicable to solar thermal energy conversion at very high temperature. Research will have to be done to lower the lattice thermal conductivity for its potential to be realized. Work will also have to be done on p-type material to achieve high doping levels since both n and p legs are needed in a thermoelectric generator.

[10] Slack, G.A., J. Appl. Phys. 35, 3460 (1964).

A brief review of other candidates for solar thermal energy conversion is given in Appendix B.

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12. Troche-Maldonado, H., "Transport Properties of Thermoelectric Si Ge Al, Si Ge B, and Si Ge P Hot-Pressed Semiconductor Alloys," Proceedings of the Second International Conference on Thermoelectricity Energy Conversion, University of Texas at Arlington, 22-24 Mar 1978.

13. Pistoulet, B., Robert, J.L., Dusseau, J.M., Roche, F.M., and Girard, P., "New Refractory Boron Silicon Semiconductor Compound with High Boron Concentration: Conduction Mechanisms and Thermoelectric Properties, 14th International Conference on Physics of Semiconductors, Edinburgh, Scotland, 4-8 Sep 1978.

APPENDIX A

PHOTON THERMAL CONDUCTIVITY

According to Devyatkova et al.[9] the thermal conductivity due to photons K_r is given by

$$K_r = \frac{16}{3} \frac{\sigma_o N^2 T^3}{\bar{\alpha}} \tag{A1}$$

where

$$\frac{1}{\alpha} = \frac{1}{51.94} \int_0^\infty \frac{x^4 \, dx}{\alpha(x)(\cosh(x)-1)} \tag{A2}$$

$$x = \frac{\hbar\omega}{kT} \tag{A3}$$

and:

n is the refractive index, σ_0 is the Stefan-Boltzmann constant ($5.67\ 10^{-9}$ mW/cm^2K^4) and σ is the absorption coefficient. This relation holds only for isotropic materials as is the case for cubic SiC. More involved formulae are given in Reference 7 for non-cubic crystals.

The optical properties of heavily doped SiC in the infrared are dominated by free carrier absorption. Since the energy gap is at least 2eV at 2000K, bound carrier absorption due to electron-hole pair production may be neglected. Lattice absorption is also negligible in comparison with free carrier absorption.

The theory of free carrier absorption gives for the real and imaginary part of the dielectric constant, ε_1 and ε_2 respectively,

$$\varepsilon_1 = \varepsilon_\infty \left(1 - \frac{\omega_p^2}{\omega^2 + \gamma^2}\right) \tag{A4}$$

(ε_∞ is optical dielectric constant)

$$\varepsilon_2 = \frac{\varepsilon_\infty \omega_p^2 \gamma}{\omega(\omega^2 + \gamma^2)} \tag{A5}$$

when

$$\omega_p^2 = \frac{4\pi Ne^2}{m^* \varepsilon_\infty} \quad (N \text{ is carrier density}) \tag{A6}$$

$$\gamma \equiv \frac{1}{\langle\tau\rangle}, \quad \langle\tau\rangle = \frac{m^* \sigma}{Ne^2} \tag{A7}$$

In terms of ε_1 and ε_2:

$$\alpha = \frac{2\omega}{c} \frac{\varepsilon_2}{2(\varepsilon' + \varepsilon_1)} \tag{A8}$$

and

$$n = \sqrt{\frac{\varepsilon_1 + \varepsilon'}{2}} \tag{A9}$$

where

$$\varepsilon' = \sqrt{\varepsilon_1^2 + \varepsilon_2^2} \tag{A10}$$

Since n is also a function of ω, we have included it inside the integral so that our more general formula is:

$$K_r = \frac{\sigma_o T^3}{9.74} \int_0^\infty \frac{n^2 x^4 \, dx}{\alpha(x) (\cosh (x)-1)} \tag{A11}$$

The value which we have used for ε_∞, the optical dielectric constant, is 6.7 from Spitzer et al.[11] The other quantities are calculable from our conduction model.

The results of our calculations show that K_r is less then 1.0 mW/cm-K as high as 2000K for carrier densities as low as 1.0×10^{20} cm^{-3}. Thus this parasitic thermal conduction is not an important limitation.

[11]Spitzer, W.C., Kleinman, D.A., Frosch, C.J., and Walsh, D.J., "Infrared Properties of SiC," reference included in SERI search, source unknown.

APPENDIX B

OTHER MATERIALS

Other materials may also be worth investigating. The Si-Ge alloys have reached ZT values of 0.8 at 1000K.[12] Materials based on Si-Ge will operate best near 1000K since the band gap is half that of SiC. They may be used in conjunction with SiC to bridge to lower temperature. A new high temperature thermoelectric p-type B_{14} Si, appears promising. A ZT value of 1.25 was reported at 1500K.[13] Higher values may be obtained at higher temperatures. However, attempts to dope B_{14} Si n-type have been unsuccessful so far. For thermoelements both n and p-type material was needed.

[12]Troche-Maldonado, H., "Transport Properties of Thermoelectric Si Ge Al, Si Ge B, and Si Ge P Hot-Pressed Semiconductor Alloys," Proceedings of the Second International Conference on Thermoelectricity Energy Conversion, University of Texas at Arlington, 22-24 Mar 1978.

[13]Pistoulet, B., Robert, J.L., Dusseau, J.M., Roche, F.M., and Girard, P., "New Refractory Boron Silicon Semiconductor Compound with High Boron Concentration: Conduction Mechanisms and Thermoelectric Properties, 14th International Conference on Physics of Semiconductors, Edinburgh, Scotland, 4-8 Sep 1978.

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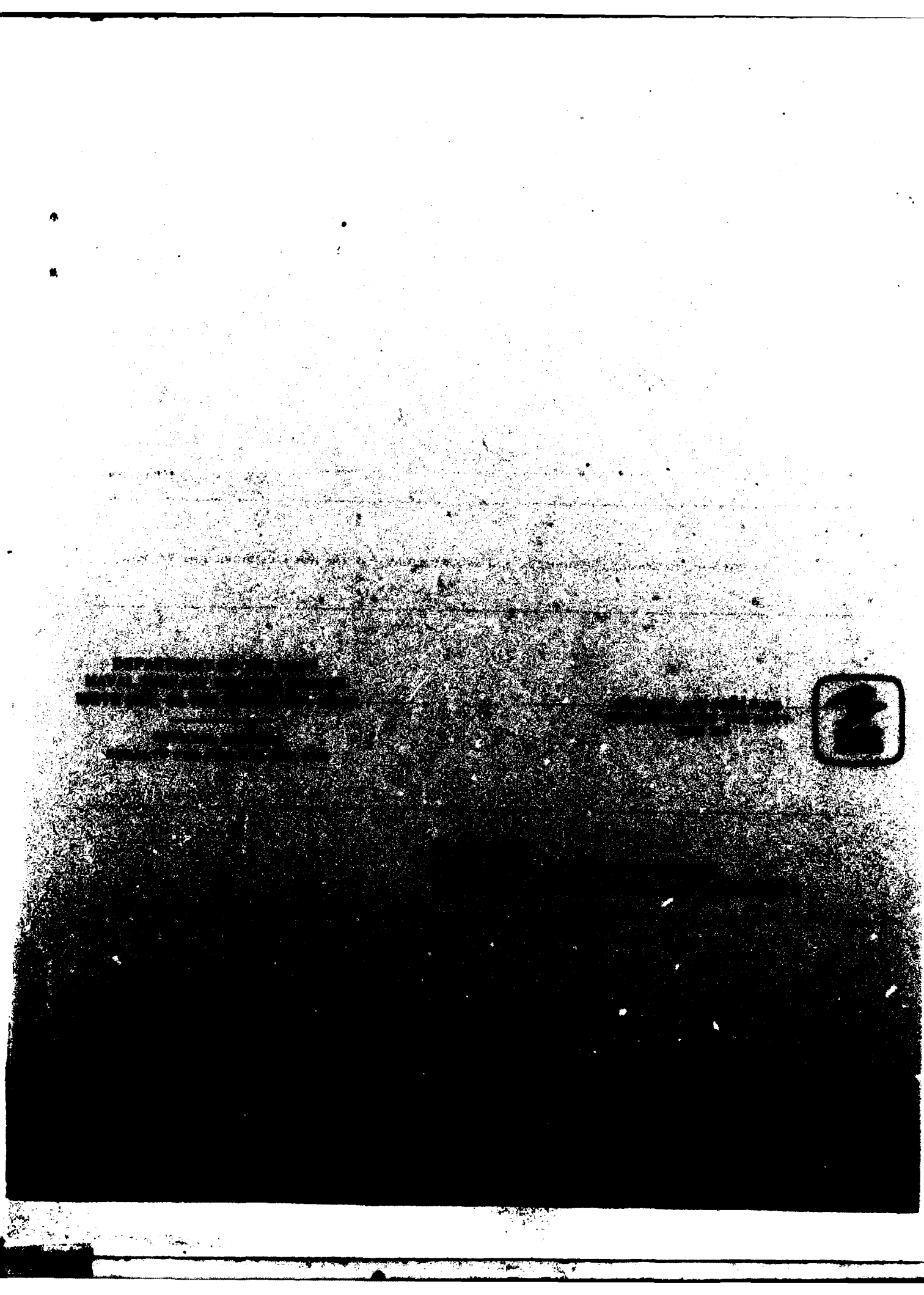


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